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Influence of roughness on the detachment force of elastic films from self-affine rough surfaces

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This article concentrates on the influence of roughness on the detachment force of elastic films on self-affine rough surfaces. It is shown that the self-affine roughness at the junction of an elastic film and a hard solid substrate influences its detachment force in a way that the detachment force can be smaller than that of a flat surface for relatively high elastic modulus E depending also on the specific roughness details. For rougher surfaces the effect of elastic energy becomes more dominant with increasing ratio between the roughness amplitude and the roughness correlation length along the interface (w/ξ). The detachment force shows a maximum after which it decreases and becomes even lower than that of a flat surface. Similar is the case of partial contact where the detachment force also increases as the contact length increases up to a maximum (for contact lengths larger than the roughness correlation length ξ), and further decreases followed by saturation. [DOI: 10.1063/1.1598636]

I. INTRODUCTION

The influence of surface roughness on the adhesion between an elastic solid and a hard solid substrate is important from both a fundamental and technological point of view, e.g., polymer/metal junctions. This topic was studied initially by Fuller and Tabor,[1] and it was shown that a relatively small surface roughness could diminish or even remove the adhesion. In their model a Gaussian distribution of asperity heights was considered with all asperities having the same radius of curvature. The contact force was obtained by applying the contact theory by Johnson, Kendall, and Roberts[2] to each individual asperity. However, this approach considers surface roughness over a single lateral length scale. The maximum pull off or detachment force is expressed as a function of a single parameter that determines (the statistically averaged) competition between compressive forces from higher asperities that try to pull the surfaces apart, and the adhesive forces from lower asperities that try to hold the surfaces together.[1]

On the other hand, random rough surfaces, which are commonly encountered for solid surfaces,[3,4] possess roughness over many different length scales rather than a single one. This case was considered by Persson and Tosatti[5] for the case random self-affine rough surfaces. It was shown that when the local fractal dimension D is larger than 2.5 the adhesive force may vanish or at least be reduced significantly. Because $D=3-H$ the roughness effect becomes more prominent for roughness exponents $H<0.5$ $(D>2.5)$. H represents the roughness exponent that characterizes the degree of surface irregularity (as H becomes smaller the surface becomes more irregular at short length scales).

These predictions[5] were limited to the case of small surface roughness and the calculations were performed using power-law approximations for the self-affine roughness spectrum which are valid for lateral roughness wavelengths $q\xi>1$ with ξ the in-plane roughness correlation length. Extension for the case of arbitrary roughness, including contributions from roughness wavelengths $q\xi<1$, were also recently performed.[6] Although the effect of various roughness parameters on the detachment force was partially analyzed, a more detailed study is necessary in order to provide a complete picture of the effect of various detailed self-affine roughness parameters.

II. RESULTS AND DISCUSSION

In the following we assume an elastic film (of elastic modulus E and Poisson's ratio ν) on top of a rough substrate. The substrate surface roughness is described by the single valued random roughness fluctuation function $h(\mathbf{r})$ with $\mathbf{r}$ the in-plane position vector $\mathbf{r}(x,y)$ such that $\langle h(\mathbf{r})\rangle=0$. The change in the total free energy of the elastic film in contact with the rough substrate is given by $U_{\mathrm{ad}}+U_{\mathrm{el}}=-A_{\mathrm{flat}}\Delta\gamma_{\mathrm{eff}}$[5] with

$$U_{\mathrm{ad}}=-\Delta\gamma A_{\mathrm{flat}}\int_0^{+\infty}du\sqrt{(1+\rho^2u)}e^{-u}$$

and

$$U_{\mathrm{el}}=A_{\mathrm{flat}}\frac{E}{4(1-\nu^2)}\int_0^{Q_c}qC(q)d^2\mathbf{q} \tag{1}$$

with $Q_c=\pi/a_0$ where a_0 is of the order of atomic dimensions, and $\Delta\gamma_{\mathrm{eff}}$ is the effective change in surface energy due to substrate roughness. We assume also a slab of thickness d that undergoes a displacement $\tilde{u}$ upon the action of a force F_{rough}. The detachment force is obtained by equalizing the elastic energy $A_{\mathrm{flat}}d(1/2)E(\tilde{u}/d)^2$ with $A_{\mathrm{flat}}\Delta\gamma_{\mathrm{eff}}$ (with A_{flat} the average macroscopic flat contact area) and taking into account the relation $F_{\mathrm{rough}}=A_{\mathrm{flat}}E(\tilde{u}/d)$ which yields[5,6]

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$$F_{\mathrm{rough}} = F_{\mathrm{flat}} (\Delta\gamma_{\mathrm{eff}}/\Delta\gamma)^{1/2} \tag{2}$$

with $F_{\mathrm{flat}} = A_{\mathrm{flat}}(2\Delta\gamma E/d)^{1/2}$ the detachment force for a flat surface. Substitution of Eq. (1) into $U_{\mathrm{ad}} + U_{\mathrm{el}} = -A_{\mathrm{flat}}\Delta\gamma_{\mathrm{eff}}$ yields $\Delta\gamma_{\mathrm{eff}}$.[6] Equation (2) is valid for constant strain field in the elastic film, which is the case for the planar geometry under consideration, and for Gaussian roughness fluctuations;[5] $\rho = \sqrt{\langle(\nabla h)^2\rangle}$ is the average local surface slope of the rough surface, and $-\Delta\gamma$ the change of the local surface free energy upon contact due to elastic film/substrate interaction.[5] For the elastic energy stored in the film we assume that the normal displacement field of the film equals $h(\mathbf{r})$.[5] $C(q)$ is the Fourier transform of the substrate height–height correlation function $C(r) = \langle h(\mathbf{r})h(0)\rangle$.

Calculations of the detachment force require knowledge of the roughness spectrum $C(q)$. For a self-affine surface roughness $C(q)$ scales as a power-law $C(q) \propto q^{-2-2H}$ if $q\xi \gg 1$, and $C(q) \propto \mathrm{const}$ if $q\xi \ll 1$.[3,4] The roughness exponent H is a measure of the degree of surface irregularity,[3,4] such that small values of H characterize more jagged or irregular surfaces at short length scales ($<\xi$). This scaling behavior is satisfied by the simple Lorentzian form[6,7]

$$C(q) = \frac{1}{2\pi} \frac{w^2\xi^2}{(1 + aq^2\xi^2)^{1+H}} \tag{3}$$

with $a = (1/2H)[1 - (1 + aQ_c^2\xi^2)^{-H}]$ if $0 < H < 1$. For other self-affine roughness correlation models see also Ref. 4 and Refs. 8, 9, and 10.

Since $C(q) \propto w^2$, the influence of the rms roughness amplitude w on F_{rough} is rather simple ($F_{\mathrm{rough}} \propto w$) while any complex dependence on the substrate surface roughness will arise solely from the roughness parameters H and ξ (or the ratio w/ξ). Equations (1)–(3) were used for the calculations in Figs. 1 and 2. Figure 1 shows that the force required to detach the film increases with increasing roughness at long wavelengths or increasing ratio w/ξ, and low values of the elastic modulus E. In this case the increment of the surface area dominates the contribution of the elastic energy. However, with increasing elastic modulus E a maximum for the detachment force is reached beyond which it starts to decrease rather fast and becomes even lower than the detachment force for a flat surface (elastic energy assisted detachment regime). Notably the maximum is more pronounced for relatively low values of the elastic modulus E so that $F_{\mathrm{rough}} > F_{\mathrm{flat}}$ over a significant range of roughness ratios w/ξ [Fig. 1(b)]. The maximum indicates that the detachment can be a *multivalued* function of the ratio w/ξ (over a limited range), which makes the interpretation of the roughness influence more complex.

Moreover, as Fig. 2(a) indicates, the detachment force shows a maximum with increasing roughness ratio w/ξ as long as $H < 0.5$. As the surface becomes smoother at short wavelengths (larger H), the detachment force reduces rather fast and it monotonically approaches the regime where film detachment is highly assisted by elastic energy or $F_{\mathrm{rough}} < F_{\mathrm{flat}}$. Alternatively the detachment force decreases with increasing H at a faster rate and magnitude for $H > 0.5$ and decreasing ratio w/ξ. The maximum that is observed for low

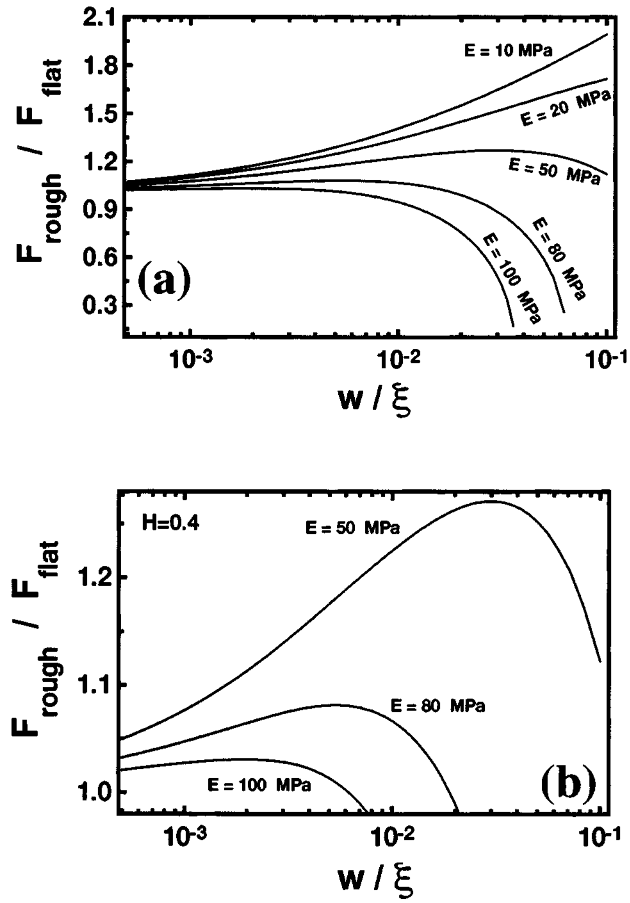

FIG. 1. (a) Detachment force $F_{\mathrm{rough}}/F_{\mathrm{flat}}$ vs roughness ratio w/ξ for roughness exponent $H = 0.4$, $w = 10$ nm, $\Delta\gamma = 4.8 \times 10^{-2}$ J/m^2, $v = 0.4$, and various elastic modulus E. (b) Detailed structure around the maximum.

roughness exponents H (< 0.5) is more pronounced for smaller long wavelength roughness ratios w/ξ.

Up to now we assumed complete contact between the elastic film and the solid substrate. If, however, only partial contact occurs at lateral length scale λ, then the real contact area $A(\lambda)$ (if the surface was smooth on all length scales shorter than λ; or apparent area of contact on the length scale λ) is related to the macroscopic nominal contact area $A(L) \cong A_{\mathrm{flat}}(\approx L^2, L \gg \xi)$ by the relation[11–13]

$$A(\lambda) = A_{\mathrm{flat}} \frac{2}{\pi} \int_0^{+\infty} \frac{\sin x}{x} e^{-x^2 G(\lambda)} dx = A_{\mathrm{flat}} \,\mathrm{erf}\left(\frac{1}{2\sqrt{G(\lambda)}}\right) \tag{4}$$

with

$$G(\lambda) = \frac{\pi}{4}\left[\frac{E}{(1-\nu^2)\sigma_0}\right]^2 \int_{2\pi/L}^{2\pi/\lambda} q^3 C(q)\, dq \tag{5}$$

and with σ_0 the applied load used to press the film onto the hard solid substrate. In this case we have for the effective detachment force due to surface roughness[6]

$$F_{\mathrm{rough}}(\lambda) = F_{\mathrm{flat}}\left[\frac{A(\lambda)}{A_{\mathrm{flat}}}\right]\left(\Delta\gamma \int_0^{+\infty} du \sqrt{(1+\rho_\lambda^2 u)}\, e^{-u} - \frac{\pi E}{2(1-\nu^2)} \int_{Q_\lambda}^{Q_c} q^2 C(q)\, dq\right)^{1/2} \tag{6}$$

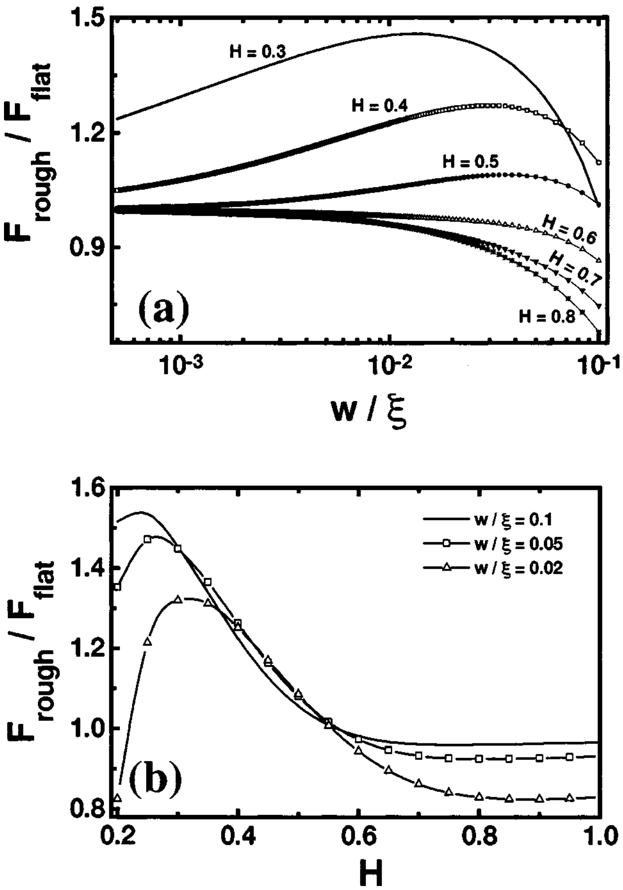

FIG. 2. (a) Detachment force $F_{\rm rough}/F_{\rm flat}$ vs roughness ratio w/ξ for various roughness exponent H, $w=10$ nm, $\Delta\gamma=4.8\times10^{-2}$ J/m^2, $v=0.4$, and elastic modulus $E=50$ MPa. (b) Detachment force $F_{\rm rough}/F_{\rm flat}$ vs roughness exponent H, $w=10$ nm, $\xi=100$ nm, $\Delta\gamma=4.8\times10^{-2}$ J/m^2, $v=0.4$, and elastic modulus $E=50$ MPa.

with $\rho_\lambda=(w/\sqrt{2}a\xi)\{[1/(1-H)][T_c^{1-H}-T_\lambda^{1-H}]+(1/H)\times[T_c^{-H}-T_\lambda^{-H}]\}^{1/2}$, $Q_\lambda=2\pi/\lambda$ and $T_\lambda=(1+aQ_\lambda^2\xi^2)$. Equations (3)–(6) were used for the calculations shown in Fig. 3.

Figure 3 indicates that the detachment force increases with increasing contact length λ. After it reaches a maximum (for $\lambda>\xi$) it further saturates for $\lambda\gg\xi$. The smaller the roughness exponent H the larger is the detachment force for contact length scales $\lambda>\xi$. The opposite occurs for small contact lengths or $\lambda<\xi$. Around the maximum area and even further to saturation we have $F(\lambda)>F_{\rm flat}$ for low roughness exponents. Figure 3(b) shows that with decreasing elastic modulus E the increment of the detachment force is more pronounced at small lateral contact length scales λ ($<\xi$). The maximum becomes more shallow and disappears depending on E. The shape of the maximum is not only affected by the elastic modulus E and the roughness exponent H, but also by the value of the lateral correlation length ξ or alternatively the ratio w/ξ. Indeed, Fig. 3(c) shows that upon smoothening of the surface the maximum broadens, preceded by a faster change of the detachment force as a function the contact length λ.

In the case of a high molecular weight monodisperse polymer it is interesting to note that the elastic modulus varies with time and the energy of adhesion depends on the time

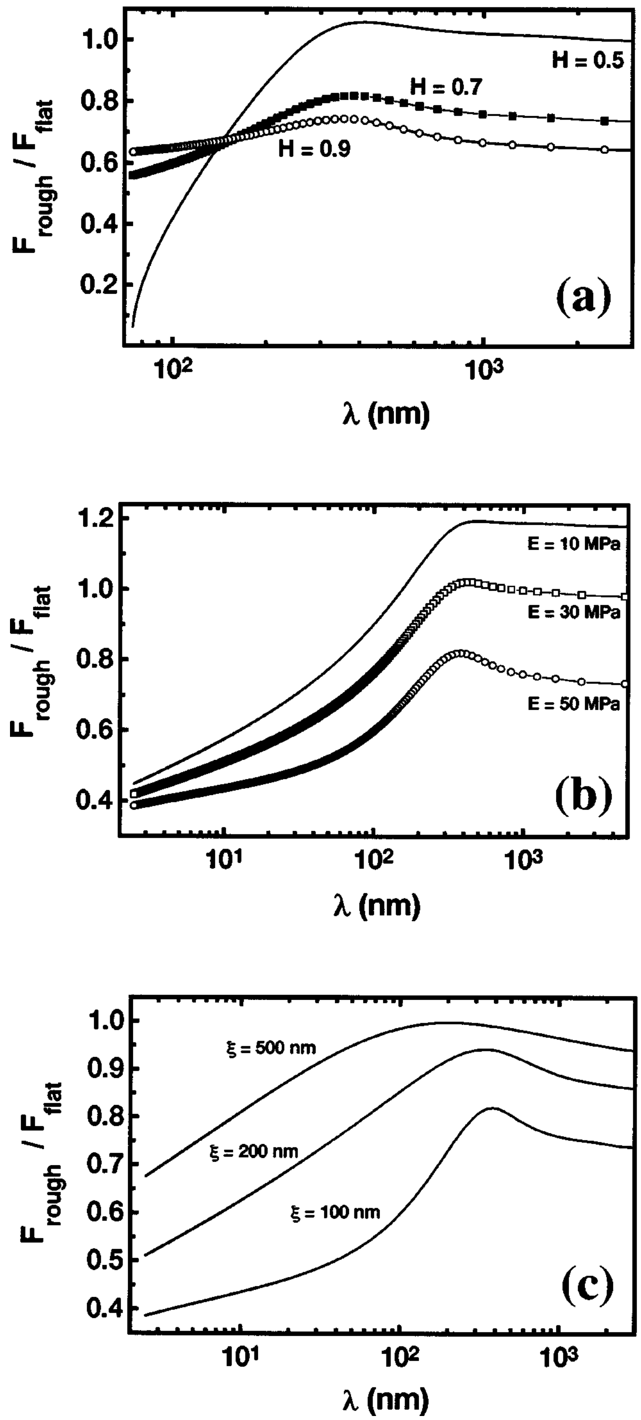

FIG. 3. (a) Detachment force vs the contact length scale λ with $\xi=100$ nm, $E=50$ MPa, and various roughness exponents H. (b) Detachment force vs the contact length scale λ with $\xi=100$ nm, various elastic modulus E, and roughness exponents $H=0.7$. (c) Detachment force vs the contact length scale λ with $H=0.7$, $E=50$ MPa, and various correlation lengths ξ. Other parameters are $w=10$ nm, $\Delta\gamma=4.8\times10^{-2}$ J/m^2, $v=0.4$, and $E/\sigma_0=50$.

of contact.[14] It is thought to decrease for small time scales ($t<\tau_c$) first, approximately according to a power law. It becomes constant till τ_d, after which it decreases again according to viscous flow. To include time dependence calculations were performed for $H=0.4$, $w=10$ nm and $\tau_c=0.005$ s and a time dependent modulus described by

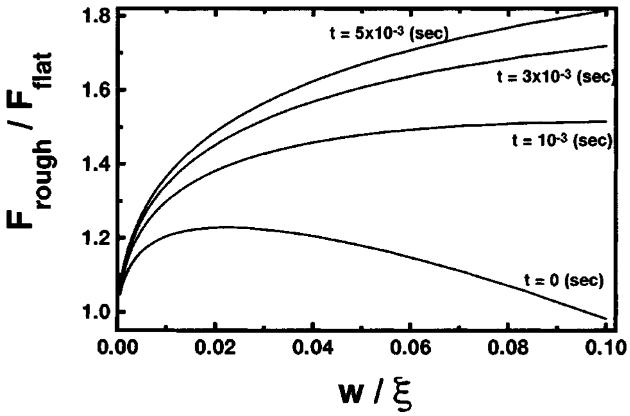

FIG. 4. Detachment force $F_{\mathrm{rough}}/F_{\mathrm{flat}}$ vs roughness ratio w/ξ for roughness exponent $H=0.4$, $w=10$ nm, $\Delta\gamma=4.8\times10^{-2}$ J/m^2, $\upsilon=0.4$, for various values of the time dependent elastic modulus.

$$E(t)=20e^{1-\sqrt{\frac{t}{\tau_c}}} \text{ with } t\leqslant\tau_c \tag{7}$$

so that $E(t=\tau_c)=20$ MPa, being a plateau modulus. The results are displayed in Fig. 4 where Eqs. (1)–(3) and (7) were used for the calculations. For $t=\tau_c$ the detachment force $F_{\mathrm{rough}}/F_{\mathrm{flat}}$ versus roughness ratio w/ξ becomes equal to the one shown in Fig. 1(a) for $E=20$ MPa. As the elastic modulus decreases with t the effect of the elastic term diminishes substantially as can be expected.

III. CONCLUSIONS

In conclusion, it is shown that the self-affine roughness at the junction of an elastic film and a hard solid substrate influences its detachment force in a way that the detachment force can be smaller than that of a flat surface for relatively high elastic modulus E depending also on the specific roughness details. When the surface becomes rougher at long wavelengths (increasing ratio w/ξ), the effect of elastic energy becomes more dominant leading to a detachment force that shows a maximum after which it decreases and becomes lower than that of a flat surface. Similar is the case of partial contact where the detachment force also increases as the contact length increases up to a maximum (for contact lengths larger than the roughness correlation length ξ), and further decreases followed by saturation. The multivalued behavior around the maximum further complicates the interpretation of the roughness influence. These results clearly indicate that the roughness has to be precisely quantified in adhesion/detachment experimental studies. However, we should note that our analytic calculations are strictly valid for elastic solids, while for real, i.e., polymers[15,16] time dependent elastic effects are present which alter besides the precise value for the elastic modulus E, also the value of $\Delta\gamma$ which is considered in the adiabatic limit. In this case surface roughness introduces fluctuating forces with a wide distribution of frequencies.[15]

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[1] K. N. G. Fuller and D. Tabor, Proc. R. Soc. London, Ser. A **345**, 327 (1975).
[2] K. L. Johnson, K. Kendall, and A. D. Roberts, Proc. R. Soc. London, Ser. A **324**, 301 (1971).
[3] P. Meakin, *Fractals, Scaling, and Growth Far from Equilibrium* (Cambridge University Press, Cambridge, 1998); J. Krim and G. Palasantzas, Int. J. Mod. Phys. B **9**, 599 (1995).
[4] Y. P. Zhao, G.-C. Wang, and T.-M. Lu, *Characterization of amorphous and crystalline rough surfaces-principles and applications, Experimental Methods in the Physical Science* (Academic, New York, 2000), Vol. 37.
[5] B. N. J. Persson and E. Tosatti, J. Chem. Phys. **115**, 3840 (2001).
[6] G. Palasantzas and J. Th. M. DeHosson, Phys. Rev. E **67**, 021604/1–6 (2003).
[7] G. Palasantzas, Phys. Rev. B **48**, 14472 (1993); **49**, 5785 (1994).
[8] G. Palasantzas and J. Krim, Phys. Rev. B **48**, 2873 (1993); G. Palasantzas, Phys. Rev. E **49**, 1740 (1994).
[9] S. K. Sinha, E. B. Sirota, S. Garoff, and H. B. Stanley, Phys. Rev. B **38**, 2297 (1988).
[10] H.-N. Yang and T.-M. Lu, Phys. Rev. B **51**, 2479 (1995).
[11] B. N. J. Persson, F. Bucher, and B. Chiaia, Phys. Rev. B **65**, 184106 (2002).
[12] B. N. J. Persson, Phys. Rev. Lett. **87**, 116101 (2001).
[13] G. Palasantzas and J. Th. M. DeHosson, J. Appl. Phys. **93**, 898 (2003).
[14] C. Creton and L. Leibler, J. Polym. Sci., Part B: Polym. Phys. **34**, 545 (1996).
[15] B. N. J. Persson, J. Chem. Phys. **115**, 3840 (2001).
[16] A. Chiche, P. Pareige, and C. Creton, C. R. Acad. Sci. Ser. IV: Phys, astrophys. **1**, 1197 (2000).